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**13. ABSTRACT** *(Maximum 200 words)*

Our efforts were aimed at the study of nonequilibrium properties of a high-density electron-hole-pair system, which was generated resonantly by femtosecond laser excitation. The investigation consisted of joint theoretical and experimental approaches. Using state of the art femtosecond experimental techniques, we studied extremely rapid physical phenomena. We have succeeded not only in completing the proposed tasks, but have also initiated some new projects and obtained very interesting results. During the last three years in this program, we have published three Physical Review Letters, nine Physical Reviews, several other papers in good journals such as Appl. Phys. Lett., J. Opt. Soc. Am., etc., nineteen invited presentations at important conferences, and several contributed papers at international conferences. The list of publications is given in the next section. Here, we summarize the highlights of our achievements during the last three years of this contract.

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# NONEQUILIBRIUM SCREENING AND EXCITON DYNAMICS PROBED BY FEMTOSECOND LASER PULSES

## FINAL REPORT

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2/15/93

U.S. ARMY RESEARCH OFFICE

DAAL03-89-K-0100

UNIVERSITY OF ARIZONA

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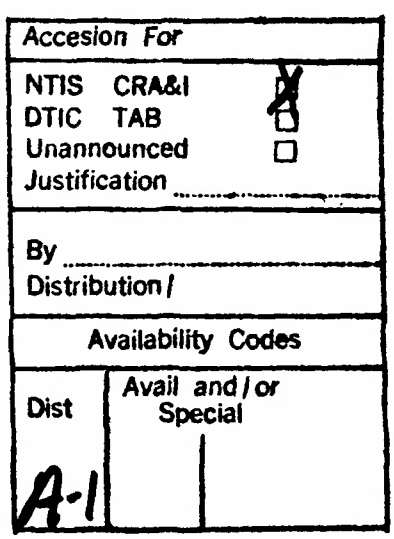


## A. STATEMENT OF THE PROBLEM STUDIED

Our efforts were aimed at the study of *nonequilibrium* properties of a high-density electron-hole-pair system, which was generated resonantly by femtosecond laser excitation. The investigation consisted of joint theoretical and experimental approaches. Using state of the art femtosecond experimental techniques, we studied extremely rapid physical phenomena. We have succeeded not only in completing the proposed tasks, but have also initiated some new projects and obtained very interesting results. During the last three years in this program, we have published three Physical Review Letters, nine Physical Reviews, several other papers in good journals such as Appl. Phys. Lett., J. Opt. Soc. Am., etc., nineteen invited presentations at important conferences, and several contributed papers at international conferences. The list of publications is given in the next section. Here, we summarize the highlights of our achievements during the last three years of this contract.

## B. SUMMARY OF RESULTS

### 1. Adiabatic Following in Semiconductors

Adiabatic following is an off-resonant effect which occurs when the duration of a light pulse is less than the phase relaxation time $T_2$ and the magnitude of the pulse detuning is greater than its inhomogeneous linewidth. Under these conditions, the quantities in the optical Bloch equations which describe the system, namely the inversion and polarization, have a time dependence determined by the instantaneous amplitude of the light-pulse envelope, i.e., they follow the field.

The above conditions on the pulse duration and detuning can be satisfied in a semiconductor using fs laser systems. This is most easily achieved for the exciton resonance in GaAs multiple-quantum-well structures (MQWs), which has a coherence time approaching one picosecond. The response of the exciton to a nonresonant light pulse can be described by the inversion and interband polarization of the system using the semiconductor Bloch equations, which include the many-body Coulomb effects in time-dependent Hartree-Fock approximation. The numerical solution of these coupled equations not only explains the Stark shift, as shown previously, but also indicates that the system's inversion adiabatically follows the field. This aspect of the ultrafast response is manifested experimentally in the time-resolved absorption measurements as a fast bleaching recovery of the exciton line.

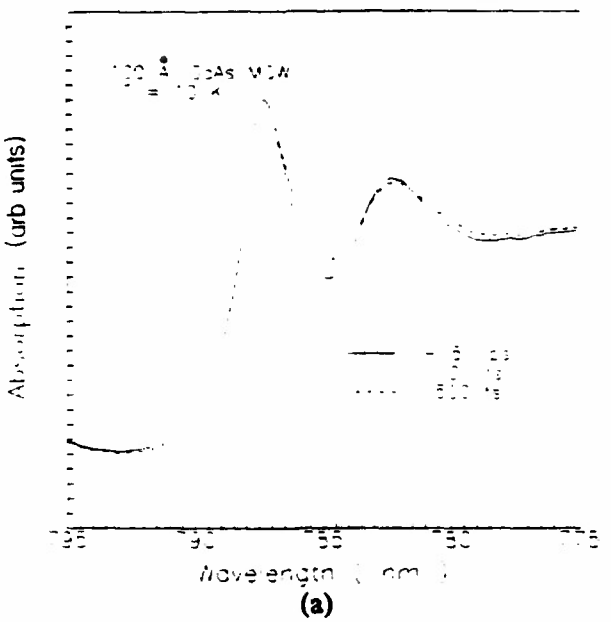
Time-resolved pump-probe experiments were carried out using a synchronously pumped mode-locked dye laser with an average output power of 25 mW, 82 MHz repetition rate, and center wavelength tunable from 850 to 870 nm for room-temperature experiments. A colliding pulse mode-locked (CPM) dye laser amplified by copper vapor lasers (CVL) in cascade operating

in 750-800 nm was employed for low-temperature ($\simeq$ 10 K) experiments. For this purpose, we first generated a continuum using CPM pulses amplified by the CVL, and then the near-IR pulses were obtained by re-amplifying the desired portion of the generated continuum using a second CVL. The autocorrelation of the pump pulse and the cross correlation of the pump and probe pulses in this case were 200 and 300 fs, respectively. The samples were molecular-beam epitaxy (MBE) grown GaAs/$Al_xGa_{1-x}As$ multiple-quantum-well and multiple-coupled-quantum-well structures (MQWs and MCQWs) with various well widths and barriers. In all room-temperature measurements the laser center wavelength was adjusted so as to be detuned between four and five $E_R$ below the heavy-hole exciton. Here $E_R$ is the bulk GaAs Rydberg energy, i.e., 4.2 meV. The detunings for low-temperature measurements were smaller. The spectral transmission of the probe through the sample for different time delays between pump and probe was measured by an optical multichannel analyzer at the output of a spectrometer.

Figure 1(a) shows the low-temperature absorption spectrum of a GaAs MCQW sample for pumping below the exciton resonance and for different time delays, $t_p$, where $t_p$ = t(probe) - t(pump). The solid curve represents the linear absorption, while the dotted spectrum corresponds to $t_p \neq 0$. It clearly shows that the heavy-hole exciton has been blue shifted and bleached. The dashed curve in Fig. 1(a), which corresponds to $t_p$ = 500 fs, demonstrates that the blue shift and bleaching are mostly recovered. The complete recovery takes nanoseconds as a result of carrier generation caused mainly by the spectral overlap of the pump and the sample's absorption spectrum. The transient exciton blue shift is the manifestation of the optical Stark effect. This bleaching recovery is the signature of the transient adiabatic following.

For the analysis of our experiments, we used the semiconductor Bloch equations, i.e., the coupled equations of motion of the expectation value of the population of the state k and the interband polarization. We solved these equations numerically for different time delays, assuming constant dephasing and carrier-relaxation rates. The calculated spectrum for comparison with the experimental data of Fig. 1(a) is shown in Fig. 1(b). As in the experiment, the pump-pulse duration and detuning satisfied the adiabatic following conditions mentioned earlier. The temporal behavior of the exciton in Fig. 1(b) shows good qualitative agreement with the data. That is, the exciton both bleaches and shifts at negative time delays. The Stark shift, which reaches a maximum at a negative time, fully recovers after several hundred fs, while the bleaching, which is maximized at $t_p$ = 0, does not quite completely recover.

Figure 2 shows the temporal behavior of the created carrier density along with the pump-pulse intensity. Note that similar to the experiments, there is a fast component of the density, which follows the pump, and a small long-lasting tail due to the incoherent component of the real carrier generation. The fast component is a result of the coherent response of the carrier density. It is the transient presence of this density which is responsible for the fast bleaching recovery. We assign this behavior to the ultrafast adiabatic following in semiconductors.



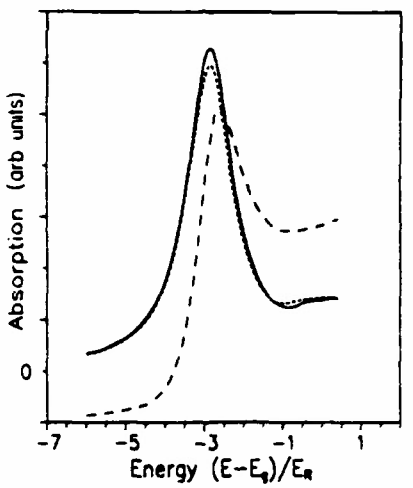




*Fig. 1. (a) Measured absorption spectra of a GaAs-AlGaAs MCQW at T = 10 K at different time delays. (b) Calculated absorption spectra for comparison with Fig. 1(a).*

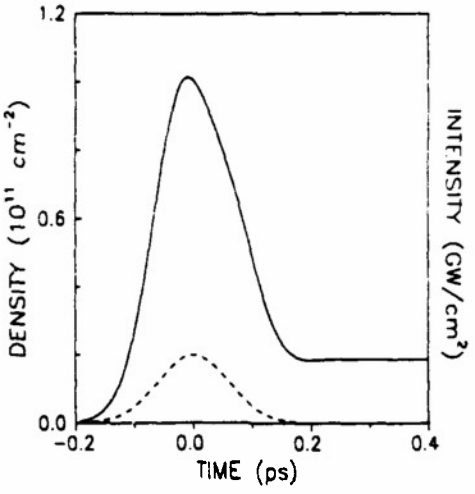



*Fig. 2. Calculated temporal behavior of the carrier density generated by pump pulse.*

## 2. Femtosecond Nonequilibrium Carrier Relaxation in Bulk CdSe

A femtosecond spectral hole-burning technique was employed to study the relaxation of nonequilibrium carriers via carrier-carrier (CCS) and carrier-LO phonon (CPS) scattering. Excitation by 70-fs laser pulses several LO phonon energies above the exciton resonance at 10 K in CdSe results in a transient spectral hole that disappears in less than 100 fs. At the onset of the pump pulse we observed a nonthermal distribution that essentially extended from the pump energy of 1.99 eV to lower energies, indicating participation of both CCS and CPS as expected. The experiments were performed with 70-fs pulses generated at 1.99 eV from an amplified colliding pulse mode-locked dye laser. Figure 3(a) shows the linear absorption spectrum of the sample at 10 K. The two excitonic peaks labeled A and B originate from the heavy-hole and light-hole valence bands split by the crystal field interaction. Absorption changes, $-\Delta\alpha$, following excitation by the pump pulse were measured as a function of time delay between the pump and probe pulses. Figure 4(a) shows $-\Delta\alpha$ spectra in 50-fs intervals. The 0-fs and 50-fs spectra show the presence of a hot nonthermal spectral hole, as indicated by the hatched area, on the high-energy side of A and B exciton bleaching. As the nonthermal distribution thermalizes, the spectral hole washes out and only the bleached excitons remain.

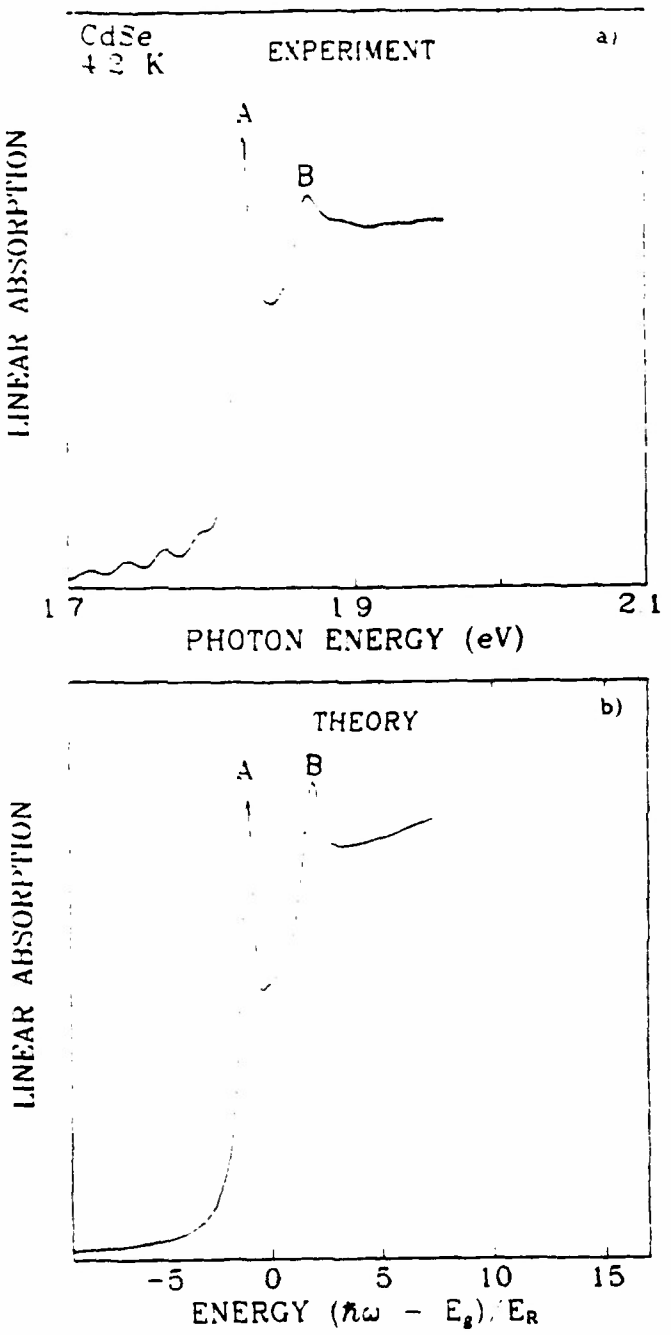



*Fig. 3. (a) The measured linear absorption of our CdSe platelet at 10 K. (b) The calculated linear absorption.*

Our theoretical analysis was based on the semiconductor Bloch equations. CCS and CPS were included in the carrier collision rates, and screening was treated quasi-statically. Results of our theory are presented in Figs. 3(b) and 4(b). Figure 3(b) shows the calculated linear absorption spectrum, consisting of the A and B excitons. The 0-fs spectrum in Fig. 4(b) clearly displays the spectral hole peaked at the pump position 12 $E_R$, where $E_R$ = 15.75 meV, the A-exciton Rydberg energy. The two excitons are completely bleached. The hole has a tail extending to

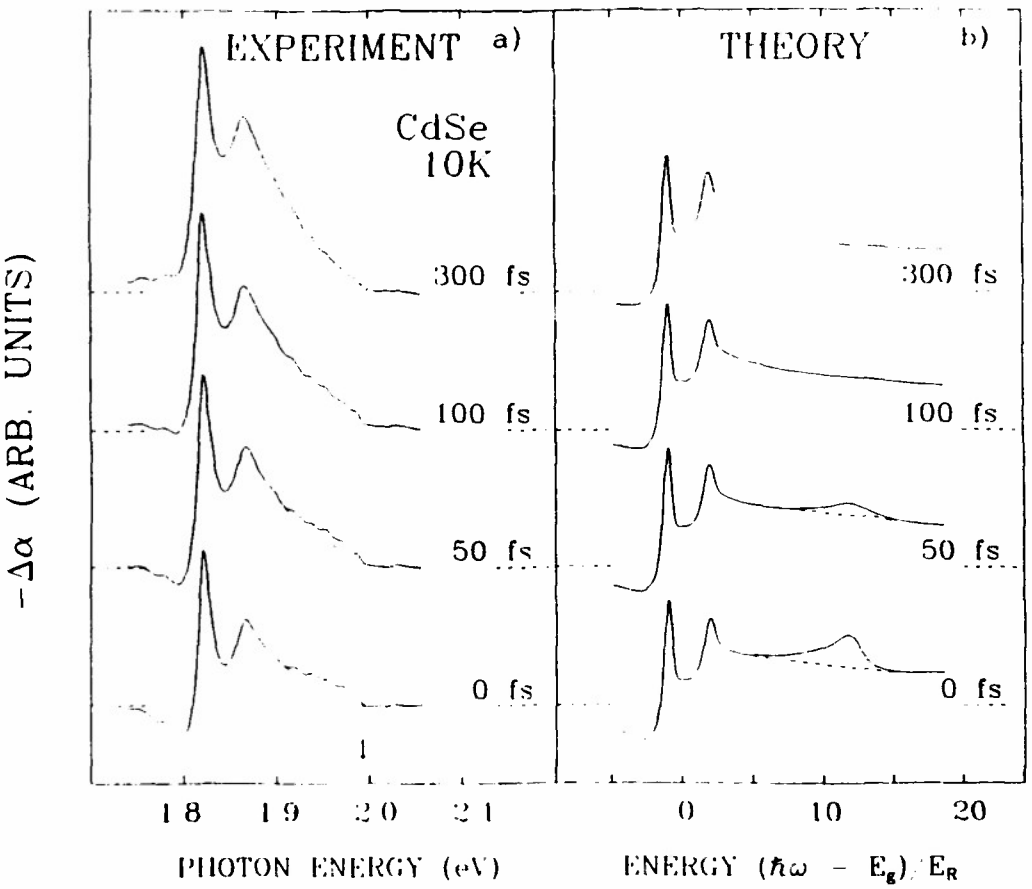



*Fig. 4. (a) The measured change in the absorption coefficient observed under the 1.99 eV excitation at 10 K. The time delay of each spectrum is shown in the figure. The hatched area shows the nonthermalized distribution of hot carriers. (b) Calculated pump-induced absorption for various time delays between the pump and probe pulses.*

low energies with respect to pump energy, as was observed in Fig. 4(a), in good agreement with the experiment. The theory also shows that the spectral hole washes out by 100 fs, but the excitons stay bleached for longer times, indicating very large carrier-carrier scattering rates. Our analysis shows that the screening by nonequilibrium carriers yields an almost instantaneous bleaching of both the exciton resonance and the Coulomb enhancement of the lower continuum states. Since the LO-phonon emission processes are essentially as fast as the CCS, the Pauli blocking (spectral hole) primarily affects the low-energy side of the pump frequency. After approximately a hundred femtoseconds, the phase-space blocking is similar to that of a high-temperature thermal plasma where, in this case, the temperature range is essentially given by the LO-phonon energy. This is a result of both CPS and CCS, where the CPS reduces the mean kinetic energy of the plasma, and the CCS yields a quasi-thermal distribution where even the

states below $\hbar\omega_{LO}$ are filled. In this investigation, the experimental results seem to be well-explained within the assumption of equilibrium phonon distributions. An enhanced temporal resolution might, however, prove the significance of nonequilibrium and/or coherent phonon effects.

### 3. Rabi Oscillations in Semiconductors

We theoretically investigated the case of exciton resonant excitation where many-body effects play a significant role in the ultrafast response of the semiconductor. It should be recalled that in the case of resonant excitation in discrete atomic systems, the dipole-coupled level populations undergo Rabi oscillations. For the case of semiconductors, we have solved the semiconductor Bloch equations for laser pulses with durations $\Delta t$, much smaller than the polarization decay ($T_2$) time; $\Delta t << T_2$. As an illustrative example, Fig. 5 shows the time dependence of the semiconductor density for a 2.2-$\pi$ pulse, i.e., $(2/\hbar)\int dt \mu E(t) = 2.2\ \pi$, exciting the system at the exciton resonance or at slightly detuned frequencies. There is no real "on-resonant" condition in semiconductors, in contrast to an atomic two-level system, since the exciton involves a large number of different k states, each of which has a slightly different detuning from the pump pulse. Different detunings imply different Rabi frequencies of the individual states, which might lead to pronounced interference effects, eventually even completely destructive interference, and the absence of Rabi oscillations. However, as Fig. 5 shows, when excited at or above the exciton resonance, the semiconductor carrier density clearly exhibits temporal oscillations. Moreover, the number of Rabi flops is basically twice that expected for a two-level system. Figure 5 also demonstrates that the amplitude of the Rabi oscillations is significantly reduced for the relatively small detuning of $-0.4E_R$ (see the long-dashed curve in Fig. 5). A careful analysis shows that the Coulomb exchange effect renormalizes the field, and thus, accounts for the doubling of the Rabi flops under the assumed conditions.

We have started experiments to verify the above predictions. A thin film of bulk CdSe was chosen as the medium to observe Rabi oscillations because the presence of a sharp exciton resonance in this material leads to the required long $T_2$ time. The calculated Rabi oscillation for the case of CdSe is shown in Fig. 6 for two values of dephasing rates. The exciton density is plotted versus the excitation field magnitude. For small dephasing rates, the density is expected to show an oscillatory behavior, while for larger dephasing rates, oscillations are expected to disappear. These Rabi oscillations were calculated for an excitation pulse of 200-fs duration. The field strength of 1 Rydberg on the horizontal axis corresponds to $\simeq 1.3\ GW/cm^2$ light intensity. These parameters are easily achievable for our femtosecond laser systems. The pump-pulse frequency was tuned inside the exciton to fulfill the resonant excitation condition. In order to monitor the exciton density, we decided to measure the exciton absorption strength as a function of laser intensity. Figure 7 shows the measured exciton absorption change as a function

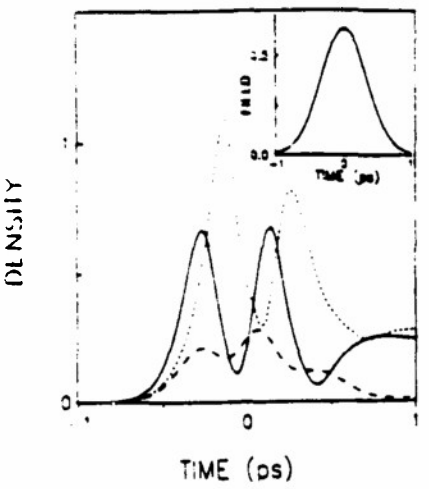



*Fig. 5. Excited carrier density in units of $a_B^{-2}$, where $a_B$ is the exciton Bohr radius for bulk GaAs for various excitation frequencies: $\hbar\omega_0 = E_g - E_R$ (solid line), $\hbar\omega_0 = E_g - 1.4E_R$ (long-dashed line), and $\hbar\omega_0 = E_g - 0.6E_R$ (short-dashed line). Inset: Pump field as a function of time for a pulse with an area of 2.2 $\pi$.*

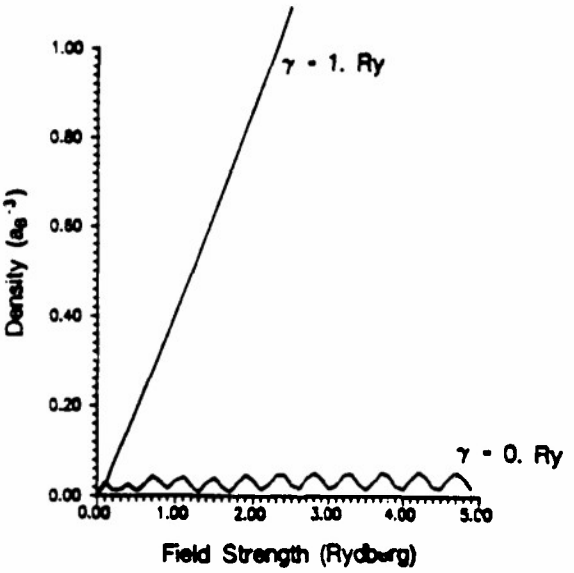



*Fig. 6. Calculated exciton density versus applied field strength (in Rydberg energy) for the case of CdSe with 200-fs laser pulse resonant excitation. The two curves correspond to two dephasing rates of $\gamma = 0$ and $\gamma = 1.0\ E_R$. The field strength of 1 $E_R$ on the horizontal axis corresponds to a laser intensity of 1.3 GW/cm².*

of pump intensity for a 150-fs delay between the pump and probe pulses. The lack of oscillations in this figure points out that exciton absorption strength is not a good monitor of the exciton density, presumably because of its quick saturation. Therefore, one needs to use a different monitor for the carrier density, such as possibly luminescence and four-wave mixing. We will discuss this subject further in the project description section.

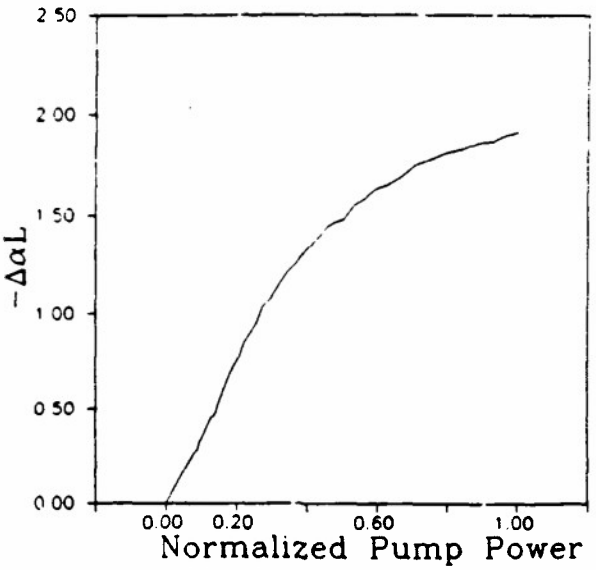



*Fig. 7. Measured exciton absorption change in a CdSe thin film at T = 10 K as a function of resonant pump intensity. The maximum power used was 0.2 GW/cm² for this case.*

We performed similar measurements on $BiI_3$, a layered semiconductor with two-dimensional excitons trapped by stacking faults. These excitons, labeled as R, S, and T, are extremely sharp, with long $T_2$ times, as shown in Fig. 8. Monitoring the absorption change at the peaks of R, S, and T excitons resulted in an interesting observation shown in Fig. 9. An oscillatory signal is detected not only at the R, S, and T peaks, but also at a wavelength of 630 nm, which is in the transparency region of the semiconductor. We verified that these oscillations are *not* the result of Rabi flopping because of the presence of the signal at 630 nm, and also because the signal lasts for $\simeq$ 30 ps. They cannot be the result of quantum beats between the excitons either because the frequency of oscillations does not correspond to the energy separation of any of the excitons. The energy separation of $\simeq$ 14.3 meV observed in these oscillations precisely matches the TO phonon frequency in $BiI_3$. Thus, we concluded that our data arise from a coherent excitation of TO phonons in the medium, presumably by impulse stimulated Raman scattering process. The $\simeq$ 30-ps decay time for the oscillations is then due to the vibrational dephasing as a result of coupling with incoherent phonons in this two-dimensional system.

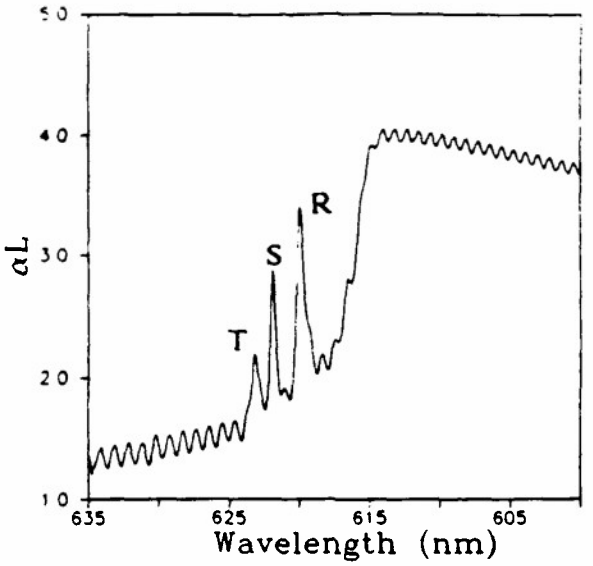



*Fig. 8. Linear absorption spectrum of $BiI_3$ at T = 10 K. R, S, and T correspond to the three stacking-fault excitons in this layered semiconductor.*

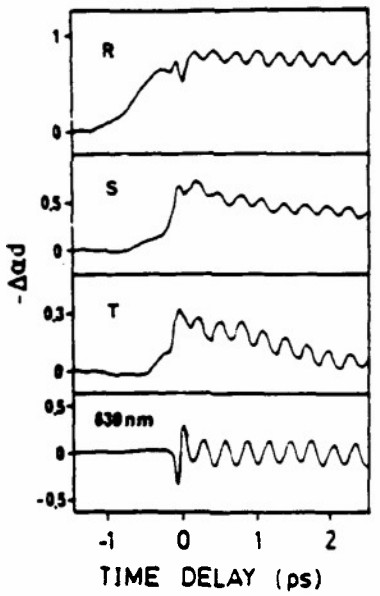



*Fig. 9. The measured change in absorption for the R, S, and T excitons and the transparency region at 630 nm of the $BiI_3$ sample of Fig. 8.*

**4. Femtosecond Dynamics of Type II Quantum Wells**

We did not originally propose this project. However, we have decided to pursue it as an additional task since it provides an interesting medium to investigate the dynamics of many-body effects associated with a one-component plasma. In the projects discussed in Sections 1 and 2 of this report, the many-body effects associated with a two-component plasma, i.e., electrons and holes, are being studied. In type-II quantum wells, on the other hand, the electrons are removed

from the holes, and the dynamics of holes or electrons can be investigated separately. For example, when the GaAs layer thickness is less than 35 Å and the AlAs layer thickness is greater than 16 Å, the lowest Γ-electron state of the GaAs well lies energetically above the X-minimum of the AlAs barrier, whereas the lowest energy hole state is still in the GaAs layer (see the inset of Fig. 10). As a result of such a band alignment, optical excitation of electron-hole pairs in the GaAs layer is followed by a spatial separation of the two plasma components; electrons scatter from the Γ-state in GaAs to the X-state in AlAs. This allows the measurement of the dynamics of the hole relaxation in the GaAs layer where they are spatially separated from the electrons in the AlAs layer. We have made such measurements at T = 10 K using 130-fs laser pulses, tuned resonantly inside the heavy-hole exciton peak of a type-II quantum-well structure consisting of ≃ 28-Å-thick GaAs (10 monolayers) and ≃ 57-Å-thick AlAs (20 monolayers) layers.

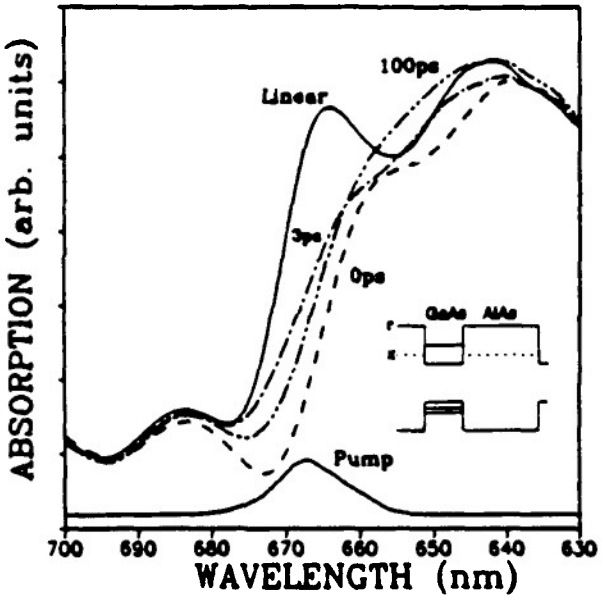



*Fig. 10. Absorption spectra at different time delays for resonant heavy-hole exciton pumping in a GaAs-AlAs type-II quantum well. The inset shows the energy level diagram for the sample.*

Figure 10 displays the absorption spectra of the sample at various time delays between the pump and probe pulses together with the pump spectrum. A bleaching and blue shift of the absorption spectrum at both the light-hole and the heavy-hole excitons is clearly observed for the zero-ps time delay. Blocking of the conduction and heavy-hole states by the electrons, which have not yet left the GaAs layer, is responsible for this effect. In the 3-ps spectrum, the light-hole exciton blue shift is completely recovered, while the heavy hole is still bleached and shifted. This behavior is caused by the fact that the Γ-X electron scattering has already taken place and only the Γ-point heavy-hole phase-space filling is left. The relaxation (cooling) of the heavy

holes contributes to the behavior of the 100-ps trace. Also, we observe a complete recovery of the light-hole exciton peak due to decreased screening of "cool" heavy holes. As the holes relax to the lowest energy states, the bleaching of the spectral region on the low-energy side of the heavy-hole exciton (see the region around $\lambda \simeq 670$ nm) is increased, while the bleaching of the high-energy side of the heavy-hole exciton (see the region around $\lambda \simeq 660$ nm) is recovered.

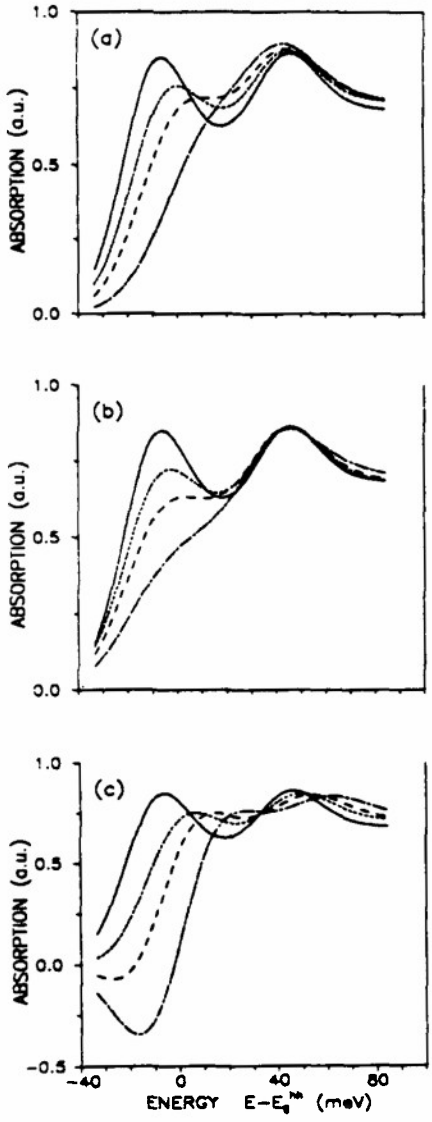



*Fig. 11. Computed absorption spectra for type-I and type-II quantum wells with GaAs well thickness of 30 Å. The spectra are for the plasma densities $na_0^2 = 0.2$ (short dashed), 0.4 (medium dashed), and 0.8 (long-short dashed). The linear spectra are shown as solid lines, and $a_0$ is the bulk-exciton Bohr radius. (a) Type-II, plasma temperature T = 30 K; (b) Type-II, T = 100 K; (c) Type-I, T = 30 K.*

In our theoretical analysis of the many-body effects in the band-edge absorption spectra of highly excited type-I and type-II semiconductor quantum-well structures, we assumed perfect electron-hole charge separation, so that only the Γ-point holes contribute to phase-space filling of the GaAs exciton states. We numerically solved the interband polarization equation, including inhomogeneous broadening due to well-width fluctuations by averaging the spectra over a distribution of well thicknesses.

Figure 11 shows computed quasi-equilibrium absorption spectra *for different plasma* densities. In the low-temperature spectra of Fig. 11(a), saturation and blue shifting of the HH exciton and a slight red shift of the LH exciton can be seen. For elevated plasma temperatures, Fig. 11(b) shows that the exciton blue shift is substantially reduced, since the holes' distributions involve more band states, substantially reducing Coulomb enhancement and Pauli blocking effects. The type-I results in Fig. 11(c) also show HH exciton saturation and blue shift, along with a blue shift of the LH exciton due to phase-space filling by the electrons.

There is no gain in the type-II spectra, whereas clear regions of negative absorption develop in the type-I spectra. This absence of gain in moderately excited ideal type-II structures is a direct consequence of the spatial-electron and hole-plasma separation. The gain part of a type-I spectrum is replaced by a zero-absorption region in type-II structures where the quasi-chemical potential coincides with the onset of absorption for sufficiently high densities.

### 5. Photon Echo in Semiconductors

We began the theoretical study of the photon echo in semiconductors using the semiconductor Bloch equations. For the photon echo case, we assumed a configuration where the two exciting pulses are incident under an angle. We took the first pulse to be weak so that we could assume linear response. The strong second pulse was treated in all orders. The center frequencies of both pulses were at the 1s exciton resonance, assuming pulse separation of $\tau = 400$ fs. Figure 12 shows that for the case of a weak first pulse, an almost instantaneous signal and no photon echo at +400 ps occurs. Our analytical calculations proved that this signal is due solely to the exchange correlation between the excited excitons. For higher pulse intensities, we see the gradual development of an echo signal, which coexists with the instantaneous signal for intermediate intensities.

To analyze the origin of this scenario, Fig. 13 plots the time dependence of the renormalized bandgap for the excitation conditions of Fig. 12. Comparing Figs. 12 and 13 reveals that the echo contribution in the time-resolved signal occurs as soon as the continuum states are shifted into resonance during the presence of the first pulse (bandgap shift below $-1E_R$ in Fig. 13). Consequently, direct continuum excitation is possible, which yields a photon echo signal at 400 fs because of the intrinsic inhomogeneous broadening of the electron-hole continuum states.

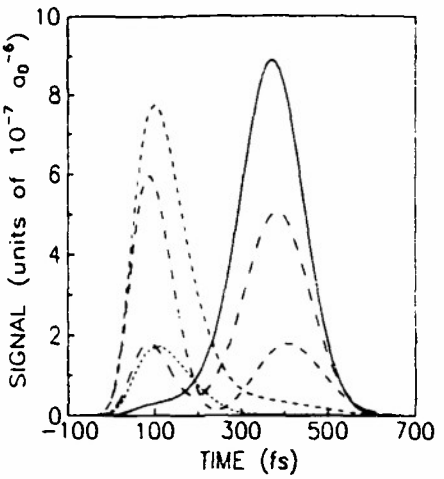



*Fig. 12. Time-resolved signal in the photon-echo direction for increasing strength of the first pulse, $E_1$. Excitation occurs at the exciton resonance. The dephasing time 200 fs, the time delay $\tau$ = 400 fs, and the pulse FWHM is 100 fs for both pulses. The peak value of the dipole coupling energy of the second pulse is $d_{cv}$ $E_1$ = 0.1 $E_R$, where the exciton binding energy $E_R$ = 16 meV in CdSe. The corresponding peak amplitudes of the first pulse are shown with increasing dash length for $d_{cv}$ $E_1$ = 0.01 $E_R$, 0.03 $E_R$, 0.05 $E_R$, 0.07 $E_R$, and for 0.1 $E_R$ (solid line).*

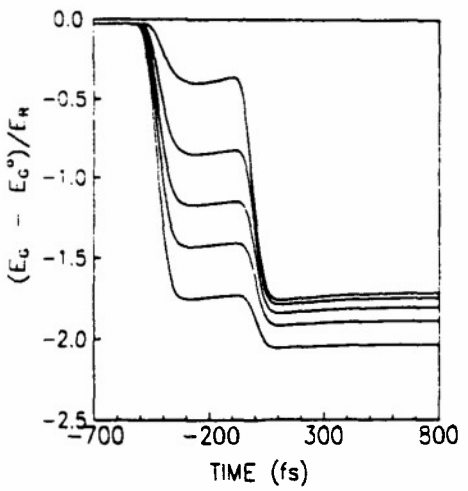



*Fig. 13. Renormalized band edge as a function time. $E_G^0$ is the unrenormalized band edge. The different curves are $d_{cv}$ $E_1$ = 0.01 $E_R$, 0.03 $E_R$, 0.05 $E_R$, 0.07 $E_R$, and 0.01 $E_R$, from top to bottom, respectively. All parameters are the same as in Fig. 12.*

## 6. Coherent Interaction of Ultrafast Pulses in Semiconductors

It is well known in atomic media that the time-dependent density of a resonantly excited two-level system exhibits oscillations, or Rabi-flops, between dipole-coupled states. We predicted earlier that this coherent phenomenon also occurs in semiconductors under resonant

femtosecond laser excitation. We have obtained preliminary experimental and theoretical results that give the first confirmations of the off-resonant Rabi-flopping in a GaAs MQW waveguide. Femtosecond laser pulses with various intensities were launched into a waveguide structure, and transmitted pulse shapes were measured using cross-correlation techniques. The results were compared with our numerical solutions of the semiconductor Bloch equations and the wave equation for the electromagnetic field.

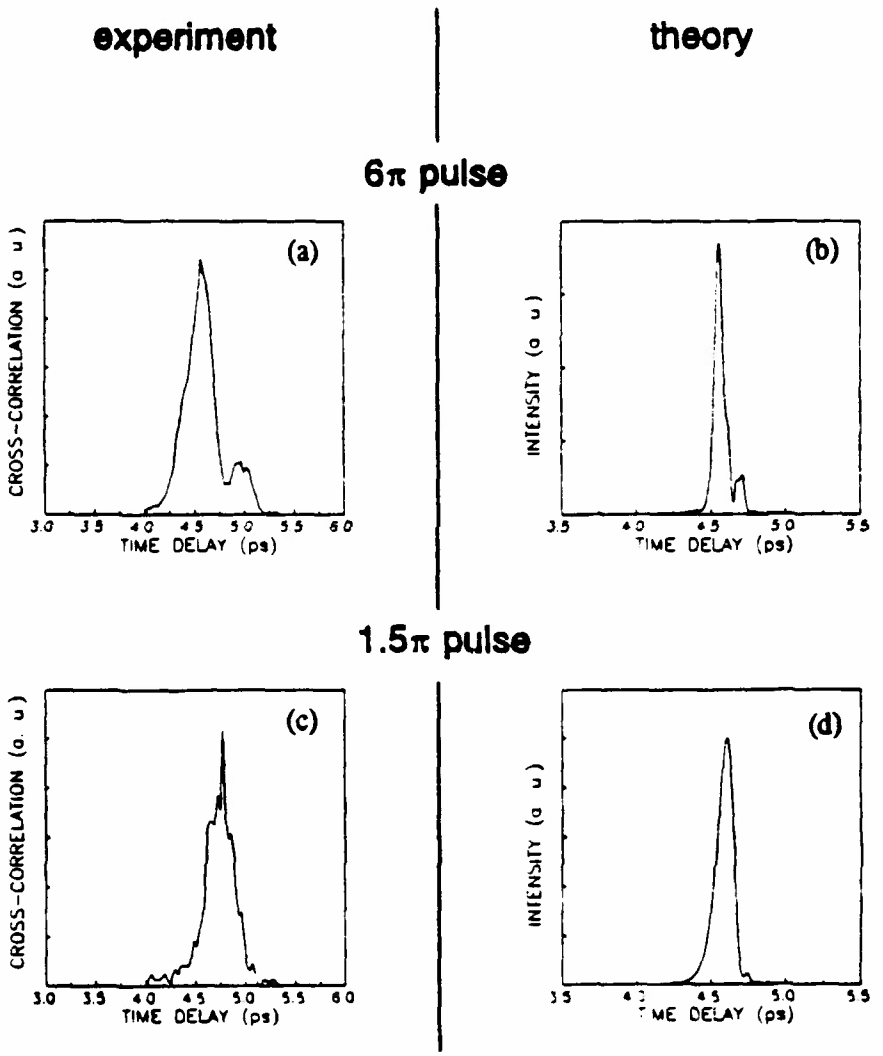



*Fig. 14. (a) and (c) are measured cross correlations of transmitted, for high and low intensity, pulses for the 0.37-mm-long sample, respectively. (b) and (d) are calculated output pulse shapes corresponding to (a) and (c) with pulse areas of 6 π and 1.5 π, respectively.*

The experimental setup was as follows: Each pulse was split into two parts. One part traveled directly to a frequency doubling crystal that was aligned for background-free second-harmonic generation, while the other part passed through a variable attenuator and the 100-Å multiple quantum well (MQW), single strip-loaded waveguide sample before it reached the doubling crystal. In Fig. 14(a) and (c) the cross-correlation signal is plotted for a 100-fs full width at half maximum (FWHM), $\lambda$ = 870 nm, pulse at two different intensities. Pulse breakup was not present at low intensities [Fig. 14(c)]; we observed two distinct peaks only at high intensities [Fig. 14(a)].

Due to the nonresonant excitation in the adiabatic following regime ($\lambda_{exciton}$ = 845 nm and $\lambda_{laser}$ = 870 nm), only few real carriers were generated. Since carriers are of minor influence under these conditions, we neglected exchange effects and treated the semiconductor in the low excitation regime. The wave equation was treated in the slowly varying envelope approximation. Figure 14(b) and (d) shows calculated cross correlations for high and low intensity inputs, respectively, with similar parameters to those in the experiment. The calculations incorporated the effects of group velocity, group velocity dispersion, and the effects of chirp and asymmetry of the input pulse.

A common feature of the experimental and theoretical results is the dip in the transmitted pulses. This is a direct result of off-resonant Rabi oscillations in the carrier density, a coherent effect. These results are therefore of great significance, since they display for the first time the effects of coherent transients on the propagation of ultrashort pulses in semiconductors.

### 7. Confinement-Induced Valence-Band Mixing in Semiconductor Quantum Dots

We have obtained experimental and theoretical results that clearly demonstrate the effects of confinement-induced valence-band mixing and Coulomb interaction in a semiconductor quantum dot sample. The experiments, which cannot be explained by a theory that assumes independent parabolic valence bands, provide strong evidence for the mixing of valence bands caused by the spherical confinement potential of the small semiconducting particles.

Quantum-confined structures are expected to lead to devices with better performances. For example, quantum dot and wire lasers and optical nonlinear devices utilizing confinement effects are considered promising for practical applications. From a fundamental point of view, these confined structures also represent an intriguing system comprising both the discrete nature of isolated elements and the band nature of large-ordered systems. The confinement of electron-hole pairs in semiconductor microstructures can result in discrete optical transitions that are usually assigned to quantum-confined states based on a simple model which assumes parabolic valence and conduction bands. However, recent theoretical work indicates that this simple model needs to be modified to account for the mixing of the valence bands caused by the spherical confining potential of the quantum dots.

In our experiment, both one- and two-photon absorption measurements were performed and compared for samples containing CdS microcrystallites in a glass matrix. An example of one- and two-photon spectra measured for a sample with an average quantum dot radius between 1 and 2 nm is shown in Fig. 15(a). It is apparent that the one- and two-photon absorption peaks occur at the same energies. Similar spectra were measured for other quantum dot samples with different average sizes. The corresponding calculated spectra in the parabolic mass approximation regime are displayed in Fig. 15(b), clearly showing the presence of a shift between the one- and two-photon absorption peaks that is not seen in the experiments. The poor agreement between the experimental spectra in Fig. 15(a) and the theoretical spectra in Fig. 15(b) shows that the independent parabolic valence-band approximation is insufficient to explain the optical transitions of CdS quantum dots.

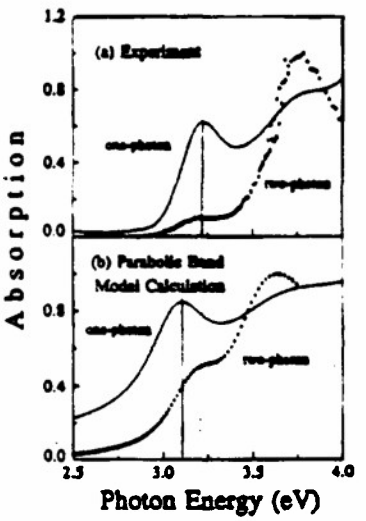



*Fig. 15. (a) Experimental results of one-photon (solid line) and two-photon (dots) absorption spectra for a quantum dot sample heat treated at 640°C for 1 hour. No observable differences are scene in the transition energies between the one- and two-photon spectra. (b) theoretical calculation of one- and two-photon absorption based on a parabolic band model.*

To improve the model, we performed a theoretical analysis which includes mixing of the heavy- and light-hole valence bands induced by the spherical confining potential in the Luttinger Hamiltonian calculations. These calculations show that the confinement-induced mixing of the valence bands results in near degeneracy of the lowest energy-hole states for the case of CdS quantum dots, which leads to the near alignment of the one- and two-photon transition energies. This can be easily seen in Fig. 16, where the calculated energies of the two lowest hole states are plotted as a function of the coupling constant for a quantum dot radius equal to half of the bulk exciton radius. In the vicinity of $\mu = 0.75$, which is appropriate for CdS, we see that the curves nearly touch, suggesting near degeneracy caused by the confinement-induced modification of the original heavy- and light-hole states.

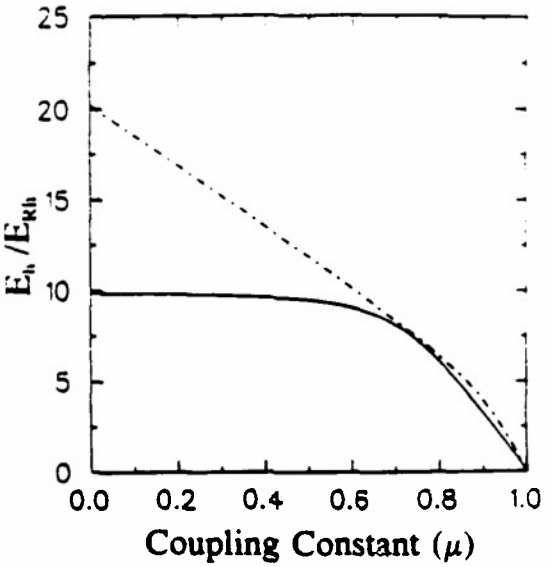



*Fig. 16. Normalized energies of the two lowest valence-band states when the mixing of the light- and heavy-hole valence bands are included in the Luttinger Hamiltonian calculation.*

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1. H. Haug and S. W. Koch, *Quantum Theory of the Optical and Electronic Properties of Semiconductors*, (World Scientific Publ., Singapore, 1990, Second Edition, 1993).
2. N. Peyghambarian, S. W. Koch, A. Mysyrowicz, *Introduction to Semiconductor Optics* (Prentice Hall, to be published Feb. 1993).

**Research Books**

1. N. Peyghambarian, ed., *Nonlinear Optical Materials and Devices for Photonic Switching*, SPIE, vol. 1216, 1990.
2. H. M. Gibbs, G. Khitrova, and N. Peyghambarian, ed. *Nonlinear Photonics* (Springer-Verlag, Berlin, 1990).

**Book Chapters**

1. N. Peyghambarian and S. W. Koch, "Semiconductor Nonlinear Materials," in *Nonlinear Photonics*, eds. H. Gibbs, G. Khitrova, and N. Peyghambarian, Springer-Verlag, 1990.

In addition to these publications, we have had nineteen invited papers and many contributed papers at international conferences.

## D. LIST OF PARTICIPATING SCIENTIFIC PERSONNEL

Paul Harten, PhD, 1992
Andy Paul, PhD, 1992
Brian Fluegel, PhD, 1992
Y. Z. Hu, PhD, 1991

In addition to these individuals, the following students participated in the program and they will get their degrees later.

Sandalphon

Ken Meissner

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**Educational background**

| | | |
|---|---|---|
| Indiana University | PhD Physics | completed-1981; degree granted-1982 |
| Indiana University | MS Physics | 1979 |
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**Employment history**

| | | |
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| University of Arizona, | | |
| Optical Sciences Center | Professor | 1991-date |
| Optical Circuitry Cooperative | Director | 1991-date |
| NTT, Japan | Visiting Professor | 7/1/90-9/1/90 |
| University of Arizona, | Associate Professor | 1988-1991 |
| Optical Sciences Center | Assistant Professor | 1985-1988 |
| | Research Assistant Professor | 1983-1985 |
| | Postdoctoral Fellow | 1982-1983 |
| Indiana University, | Postdoctoral Fellow | 1981-1982 |
| Physics Department | Research Assistant | 1977-1981 |

**Fields of major current interest**

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**Professional Activities**

--Topical Editor of Optics Letters, 1992-1995 (for the nonlinear optics section).
--Program Chair of the Interdisciplinary Laser Science (ILS) Conference, Toronto, 1993.
--Member of the program committee of Conference on Lasers and Electro-optics (CLEO), subcommittee on Ultrafast Optics and Optoelectronics, Baltimore, Maryland, May 1993.

--Vice Chair of the "Ultrafast Phenomena" section of the OSA Annual Meeting - 1992
--Vice Program Chair and Chair of the subcommittee on "Nonlinear Optics and Ultrafast Phenomena" of the ILS Conference, Albuquerque, New Mexico, Sept. 21-26, 1992.
--Member of the Editorial Board of the "Nonlinear Optics" journal, Nov. 1991-present.
--Member of the Program Committee of the International Quantum Electronic Conference, IQEC'92, Vienna, Austria, June 14-19, 1992.
--U.S. member of the Program Committee of the Third International Workshop on Nonlinear Optics and Excitation Kinetics in Semiconductors, May 18-21, 1992, Bonn-Bad Honnef, Germany.
--Chairman of the symposium on "Nonlinear Optics" of the Electrochemical Society Meeting, Oct. 11-16, 1992, Toronto, Canada.
--Chairman of the subcommittee on "Applications of Nonlinear Optics and Laser Spectroscopy," Conference on Lasers and Electro-Optics (CLEO), Baltimore, Maryland, May 13-17, 1991.
--Member of the Program Committee of the Quantum Electronics and Laser Science Conference (QELS), Baltimore, Maryland, May 11-17, 1991.
--Chairman of the Subcommittee on "Nonlinear Optics and Ultrafast Phenomena" of the International Laser Science Conference (organized by the American Physical Society), Monterey, California, Sept. 22-26, 1991.
--Member of the Program Committee of the SPIE Conference on Nonlinear Optics and Materials, Jan. 19-24, 1992, Los Angeles, California.
--Member of the Steering Committee of the Photonic Research Center at the United States Military Academy, West Point, 1989-present.
--Member of the Program Committee of SPIE Conference on *Devices for Optical Processing*, July, 1991, San Diego, California.
--Member of the Advisory Committee of the Conference on Nonlinear Dynamics of Optical Systems, Afton, Oklahoma, June 4-8, 1990.
--Chairman of SPIE Conference on *Nonlinear Optical Materials and Devices for Photonic Switching*, Los Angeles, CA, January 16-17, 1990.
--U.S. Chairman of *Optical Bistability IV* Conference, Aussois, France, March 1988.
--Member of the Program Committee of the International Quantum Electronics Conference (IQEC), Anaheim, California, May 21-25, 1990.
--Member of the Program Committee of CLEO, Anaheim, California, May 21-25, 1990.
--Member of the Program Committee of QELS Conference, Baltimore, MD, 1989.
--Member of the Program Committee of CLEO, 1989, Baltimore, MD.
--Member of the Program Committee of the IQEC, Baltimore, Maryland, April 28-May 1, 1987.
--Chairman of SPIE Conference on *Optical Computing and Nonlinear Materials*, Los Angeles, CA, Jan. 11-13, 1987.
--Cochairman of NSF Meeting on *Light Wave Technologies*, Tucson, Arizona, May 22-23, 1986.
--U.S. Program Chairman of *Optical Bistability 3* Conference, Tucson, Arizona, December 2-4, 1985.
--Cochairman of SPIE Conference on *Digital Optical Computing*, Los Angeles, California, January 11-17, 1987.
--Member of the Program Committee of CLEO, Baltimore, Maryland, April 28-May 1, 1987.
--Member of the Program Committee of CLEO, Anaheim, CA, April 25-29, 1988.
--Member of the Organization and Program Committee of the International Meeting on "Optical Nonlinearity of Semiconductors," Berlin, German Democratic Republic, Aug. 22-25, 1988.
--Session Chairman of many Conferences including, 1987 Gordon Conference on Nonlinear Optics and Lasers (Wolfboro, New Hampshire), CLEO 1988, CLEO 1987, IQEC 1988, IQEC 1987, QELS 1989, CLEO 1989, OSA Annual Meeting 1988, etc.

--Member of Coordinating Committee of the Optical Society of America, 1985.
--Member of Review Panel of the National Science Foundation, 1986, 1989.
--Guest Editor of special issue of Optical Engineering on Optical Computing, January 1987.
--Referee, National Science Foundation, Physical Review Letters, Physical Review B, Optics Letters, Applied Physics Letters, Journal of the Optical Society of America, Applied Optics, IEEE Journal of Quantum Electronics, Optics Communications, Electronics Letters.
--Member of the Graduate College Committee on Graduate Studies at the University of Arizona.
--Member of the PhD preliminary examination committee of the Optical Sciences Center.

**Honors**

--TRW Young Faculty Award, 1989-1990
--3M Company's Young Faculty Award, 1987-1988 and 1988-1989.
--Outstanding graduate student in research, Physics Department, Indiana University, 1981.

**Consulting**

--US Army Advanced Research & Development Command, 1986-1987
--Honeywell Corporation, 1985-present
--Celenese Company, 1988-1989

**Professional Society Affiliations**

--American Physical Society
--Optical Society of America
--SPIE, Society of Optical Engineers

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1. S.W. Koch, *Dynamics of First-Order Phase Transitions in Equilibrium and Nonequilibrium Systems*, Springer Lecture Notes in Physics 207, Springer Verlag, Berlin (1984).

2. H. Haug and S.W. Koch, *Quantum Theory of the Electronic and Optical Properties of Semiconductors*, World Scientific Publ., Singapore (1990); second revised edition (1993).

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**Invited Talks and Publications in Conference Proceedings:**

1. H. Haug and S.W. Koch, "Nonequilibrium Phase Transitions in Highly Excited Semiconductors," International Symposium on Synergetics, Bielefeld, Fed. Rep. Germany, September 24 - 29, 1979. Published in: *Dynamics of Synergetic Systems*, *p.* 57, *ed.* H. Haken, Springer Verlag, Berlin (1980).

2. H. Haug, S.W. Koch, R. März, and S. Schmitt-Rink, "Optical Nonlinearity and Bistability in Semiconductors due to Biexciton Formation," International Conference on Luminescence, Berlin, Fed. Rep. Germany, 1981.

3. R.C. Desai, S.W. Koch, and F.F. Abraham, "Growth-Law Dynamics in a Phase Separating Fluid," Conference on Nonlinear Fluid Behavior, Boulder, CO, June 7 - 11, 1982.

4. S.W. Koch, F.F. Abraham, and R.C. Desai, "Numerical Simulation of Spinodal Decomposition in Simple Fluids," Conference on Kinetics of Phase Change, St. Barbara, CA, March 1 - 5, 1982.

5. S.W. Koch, "Molecular Dynamics Simulation of Phase Transitions in Physisorbed Noble Gases," 3rd General Conference of the Condensed Matter Division of the EPS, Lausanne, Switzerland, March 28 - 30, 1983.

6. S.W. Koch, "Molecular Dynamics Simulations of Phase Transitions in Noble Gas Overlayers," Solid State Physics Conference, Oxford, Great Britain, December 14 - 16, 1983.

7. S.W. Koch and H. Haug, "Theory of Resonance Enhanced Optical Nonlinearities and Bistability in Semiconductors" (in German), Spring Meeting of the German Physical Society DPG, Giessen, Fed. Rep. Germany, March 19 - 23, 1984.

8. H. Haug, S.W. Koch, H.E. Schmidt, and M. Lindberg, "Soliton Structures in the Excitation due to Induced Absorption," International Conference on Instabilities and Dynamics of Lasers and Nonlinear Optical Systems, Rochester, NY, 1985.

9. S.W. Koch, H. Haug, and M. Lindberg, "Optical Nonlinearities and Instabilities in Semiconductors," 6th General Conference of the Condensed Matter Division of the EPS, Stockholm, Sweden, March 22 - 25, 1986.

10. H.M. Gibbs, N. Peyghambarian, Y. H. Lee, M. Warren, A. Chavez-Pirson, S.W. Koch, A. C. Gossard, and W. Wiegmann, "Room-Temperature Bulk GaAs: Dominant Nonlinearities, Fast-Recovery Gates, Arrays for Parallel Processing," NSF Workshop on Optical Nonlinearities, Fast Phenomena and Signal Processing, Tucson, AZ, May 22 - 23, 1986 (*p.* 51 in Workshop Proceedings, *ed.* N. Peyghambarian).

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12. S.W. Koch, "Dynamics and Evolution of Structure During Phase Transitions on Surfaces," International Meeting on Advances in Phase Transitions and Disorder Phenomena, Amalfi, Italy, June 25 - 27, 1986. Published in: *Advances on Phase Transitions and Disorder Phenomena*, p. 72, eds. G. Busiello, L. DeCesare, F. Mancini, and M. Marinaro, World Scientific Publ., Singapore (1987).

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66. S.W. Koch, Y.Z. Hu, and R. Binder, "Exchange Effects and Multi-Wave Mixing in Quantum-Confined Semiconductors," NATO Advanced Research Workshop *Physics of Few-Electron Nanostructures*, Noordwijk aan Zee, Netherlands, Sept. 23 - 26, 1992.

67. S.W. Koch, "Theory of Linear and Nonlinear Optical Properties of Semiconductor Quantum Dots," 182nd Meeting of the Electrochemical Society, Toronto, Canada, Oct. 11 - 16, 1992.

68. S.W. Koch, "Microscopic Theory of Semiconductor Microlasers," March Meeting of the APS, Seattle, Washington, March 22 - 26, 1993.

69. S.W. Koch, Institute of Physics Annual Conference, Brighton, UK, April 19 - 22, 1993.

70. S.W. Koch and R. Binder, "Optical Nonlinearities in Quantum Confined Semiconductors," workshop on Optical Properties of Mesoscopic Semiconductor Structures, Snowbird, Utah, April 20 - 23, 1993.

71. S.W. Koch, "Coherence and Intra-Band Interaction in *D*-Dimensional Semiconductors," Workshop *Semiconductor Optics*, Marubrg, Germany, May 14 - 18, 1993.

72. W.W. Chow and S.W. Koch, "The Use of the Semiconductor Gain Medium as an Experimental Variable in the Study of Laser Instabilities," SPIE Annual Meeting, San Diego, CA, July 11- 16, 1993.

73. S.W. Koch, Nato Advanced Research Workshop on Coherent Optical Interactions in Semiconductors, Cambridge, UK, August 11 - 14, 1993.

74. S.W. Koch, "Femtosecond Coherent Effects in Semiconductors," ILS/OSA Meeting, Toronto, Canada, 1993.

75. S.W. Koch, "Coherent Effects in Semiconductors," $VI^{th}$ International Symposium on Quantum Optics, Rotorua, New Zealand, Jan. 24 - 28, 1994.

**Contributed Talks and Publications in Conference Proceedings:**

1. S.W. Koch and H. Haug, "On the Theory of Stimulated Emission in Highly Excited Semiconductors" (in German), Spring Meeting of the German Physical Society DPG, Münster, FRG, 1977.

2. C. Klingshirn, W. Maier, B. Hönerlage, H. Haug, and S.W. Koch, "Quantitative Investigations on the Recombination Involving Free Particle Scattering Processes in Highly Excited Blende Type II-VI Compounds," International Conference on Recombination in Semiconductors, Southampton, Great Britain, 1978.

3. S.W. Koch and H. Haug, "On the Electron-Hole Droplet Nucleation in Highly Excited Semiconductors" (in German), Spring Meeting of the German Physical Society DPG, Münster, FRG, 1979.

4. S.W. Koch, "On the Electron-Hole-Droplet Nucleation in Indirect and Direct-Gap Semiconductors" (in German), DPG-Round Table Discussion on Excitonic Polaritons and Highly Excited Semiconductors, Reisensburg/Ulm, FRG, 1979.

5. S.W. Koch, "On the Electron-Hole-Plasma Phase Transition in Highly Excited Direct-Gap Semiconductors," 1980 Annual Conference of the Condensed Matter Division of the EPS, Antwerpen, Belgium, 1980. Published as: S.W. Koch, "On the Hydrodynamics of the Electron-Hole Plasma Phase Transition in Highly Excited Semiconductors," in: *Recent Developments in Condensed Matter Physics. Vol. 3, p.* 249, Plenum Publ., New York (1981).

6. J.P. Löwenau and S.W. Koch, "Formation and Decay of Electron-Hole-Droplets in Highly Excited Germanium" (in German), Spring Meeting of the German Physical Society DPG, Münster, FRG, 1981.

7. R. März, H. Haug, S.W. Koch, and S. Schmitt-Rink, "Nonlinearity of the Dielectric Function due to Formation of Biexcitons and Optical Bistability" (in German), Spring Meeting of the German Physical Society DPG, Münster, FRG, 1981.

8. H. Haug, S.W. Koch, R. März, and S. Schmitt-Rink, "Optical Nonlinearity and Instability in Semiconductors due to Biexciton Formation," International Conference on Excited States and Multiresonant Nonlinear Optical Processes in Solids, Aussois, France, March 18 - 20, 1981.

9. S.W. Koch, R.C. Desai, and F.F. Abraham, "Spinodal Decomposition of a One-Component Fluid: Hydrodynamics," 9th Midwest Solid State Theory Symposium, Argonne, IL, November 2 - 3, 1981.

10. H.E. Schmidt, R. Neumann, H. Haug, and S.W. Koch, "Influence of Noise on the Optical Bistability of a Two-Photon Resonance" (in German), Spring Meeting of the German Physical Society DPG, Freudenstadt, FRG, 1983.

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12. H.E. Schmidt, H. Haug, and S.W. Koch, "Absorptive Optical Bistability in Semiconductors due to Band-Gap Reduction" (in German), Spring Meeting of the German Physical Society DPG, Giessen, FRG, 1984.

13. S.W. Koch, H.E. Schmidt, and H. Haug, "Theory of Absorptive Bistability," 3rd Trieste ICTP-IUPAP Semiconductor Symposium on High Excitation and Short Pulse Phenomena, Trieste, Italy, July 2 - 6, 1984.

14. C. Ell, S. Schmitt-Rink, S.W. Koch, H.E. Schmidt, and H. Haug, "Theory of Optical Properties of Semiconductor Quantum-Well Structures," 5th General Conference of the Condensed Matter Division of the EPS, Berlin, Fed. Rep. Germany, March 18 - 22, 1985.

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22. M. Lindberg, S.W. Koch, and H. Haug, "Self-Excited Oscillations of an Induced Absorber in a Resonator," Spring Meeting of the Quantum Optics Division of the German Physical Society DPG, Heidelberg, FRG, 1986.

23. G.R. Olbright, N. Peyghambarian, S.W. Koch, and L. Banyai, "Experimental and Theoretical Investigations of the Origin of Optical Nonlinearities of Semiconductor-Doped Glasses," IQEC'86 XIV International Quantum Electronics Conference, San Francisco, CA, June 1986.

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25. N. Peyghambarian, G.R. Olbright, B.D. Fluegel, and S.W. Koch, "Femtosecond Dynamics of Bandgap Renormalization and Bandfilling in a CdSe-Microcrystallite-Doped Glass," postdeadline paper PD-20, IQEC'86 XIV International Quantum Electronics Conference , San Francisco, CA, June 1986.

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